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Nonlinear optical processes in one-dimensional polymers

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ABSTRACT

New phenomena are observed theoretically in nonlinear optical responses of polydiacetylene-toluene sulfonate (PTS) irradiated by a strong laser beam. Within a two-level model of PTS, the transient behavior of the induced susceptibility is investigated. Optical nutation is found, and the optical Stark blue shift and bleaching are also found in qualitative agreement with experiments. In the steady state, a new type of optical tristability mediated by phonons is found. A possible mechanism responsible for this tristability is discussed.

1. INTRODUCTION

Nonlinear optical processes have been employed to realize light frequency conversion, light modulation and switching, optical logic and memory storage in optical computing, signal processing and image analysis.¹ Because of such immense potential applications, it has been one of the most active areas of research for both basic understanding of the mechanism and technological development of electro-optical or all-optical devices for communication and computer systems.

Organic polymers are known to exhibit giant nonlinear optical susceptibilities with fast response time.² In particular, the third-order susceptibility $\chi^{(3)}$ of conjugated polymers has attracted much attention. It is expected to play an important role in phase conjugation, optical bistability and all-optical modulation.

The large optical nonlinearities in conjugated polymers, first predicted over ten years ago,³ have been observed in numerous systems in recent years. The largest $\chi^{(3)}$ is recorded for trans-polyacetylene⁴ (CH)_x for which a number of theoretical investigations have been carried out very recently. On the basis of a one-dimensional model,^{5,6} $\chi^{(3)}$ has been calculated for (CH)_x by means of the Keldysh Green's function,⁷ and for pristine (CH)_x with neutral solitons.⁸ Furthermore, the chain-length effects on $\chi^{(3)}$ for (CH)_x have also been studied.

Meanwhile, nonlinear optical responses in polydiacetylene (PDA) have attracted much attention. The nonlinear refraction index n_2 of PTS films¹⁰ and PDA-4BCMU¹¹ have been measured, and phonon-mediated excitonic processes in PTS have been observed in transient nonlinear optical response^{12,13} and in an optical Stark effect experiment.¹⁴

We report, in this paper, theoretical studies of the transient nonlinear optical response of virtual excitons in PTS and a new type of optical tristability of PTS in an optical cavity.

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2. TRANSIENT OPTICAL RESPONSE OF PTS

The investigation of coherent nonlinear optical processes caused by semiconductor excitations in the transparent region well below the absorption edge^{15,18} has been extended to polymers.¹²⁻¹⁴ It is known that virtual excitons are created when the frequency of an incident laser beam is tuned below the exciton resonance. Such excitonic effects can be explored more naturally by probing the susceptibility¹⁸ and looking for transient changes that persist as long as the excitation. Provided that the sample is strongly excited well below the conduction band edge, the real part of the susceptibility can be examined by measuring the index of refraction, and the imaginary part is explored by the absorption spectrum.

As PDA (and, in turn, PTS) possesses fairly large $\chi^{(3)}$ and small transmission loss α , which can be reduced to as low as¹⁹ $\alpha < 1$ db/cm, PDA can have a rather large ratio $\chi^{(3)}/\alpha$. In addition, PDA is relatively easy to use in the construction of waveguides. In this section we take PTS as an example and study the transient behavior of its induced optical susceptibility due to irradiation by a strong pumping laser and a weak probing laser. The two lasers may have pulse duration of femtoseconds to picoseconds. For the PTS system we adopt a two-level model to describe its ground state and exciton state. The lifetime of the exciton is also introduced phenomenologically.

2.1 Two-level model

The two states of PTS are denoted by $|+\rangle$ and $|-\rangle$. We assume a dipole transition and define the dipole operators

$$S^- = |-\rangle\langle +|, \quad (1a)$$

$$S^+ = |+\rangle\langle -|. \quad (1b)$$

The inversion operator is given by

$$S^z = \frac{1}{2} (|+\rangle\langle +| - |-\rangle\langle -|). \quad (2)$$

For simplicity, the effect of phonon mediation²² is ignored here. (Its inclusion will be discussed and published elsewhere.) Thus we consider a single virtual exciton and write the Hamiltonian in the rotating-wave approximation as

$$H = H_0 + H' \quad (3a)$$

$$H_0 = \hbar\omega_x S^z - \frac{1}{2} (\mu_0 E_p e^{i\omega_p t} S^- + \text{h.c.}) \quad (3b)$$

$$H' = -\frac{1}{2} (\mu_0 E_t e^{i\omega_t t} S^- + \text{h.c.}), \quad (3c)$$

where ω_x is the exciton frequency, μ_0 is the dipole matrix element, and E_p and E_t are the amplitude and frequency of the pump and test fields, respectively. Here we have neglected the momentum dependence of virtual excitons.^{13,23} This Hamiltonian uses dipole or projection operators to describe the dipole transition between the ground state and the excitonic state of the material by external fields.

It is different from that of Ref. 13, where creation and annihilation operators were used for virtual excitons.

We assume further that initially the system is in its ground state with no virtual exciton present. Thus we have $|\psi(0)\rangle = |-\rangle$ at $t = 0$. For any later time $t > 0$, the state can be represented by

$$|\psi(t)\rangle = C_+(t)|+\rangle + C_-(t)|-\rangle \quad (4)$$

Plugging (4) into the Schrödinger equation with the Hamiltonian (3), we find the equations satisfied by the amplitudes C_{\pm} in the rotating frame as

$$\dot{C}_+(t) = i\Omega_p^* e^{-i\Delta_p t} C_- + i\Omega_t^* e^{-i\Delta_t t} C_- - \gamma C_+ \quad (5a)$$

$$\dot{C}_-(t) = i\Omega_p e^{i\Delta_p t} C_+ + i\Omega_t e^{i\Delta_t t} C_+ , \quad (5b)$$

where we have defined $\Omega_{p,t} = \mu_o E_{p,t}/2\hbar$ and $\Delta_{p,t} = \omega_{p,t} - \omega_x$. In addition, a damping rate γ has been introduced to describe the very short lifetime of the virtual excitations.

Since the test field is in most cases much weaker than the pump field, we can safely apply perturbation theory to solve (5). The zeroth-order equations are obtained by setting $E_t = 0$ in (5), namely,

$$\dot{C}_+^o = i\Omega_p^* e^{-i\Delta_p t} C_-^o - \gamma C_+^o \quad (6a)$$

$$\dot{C}_-^o = i\Omega_p e^{i\Delta_p t} C_+^o \quad (6b)$$

In what follows, we assume for simplicity $\mu_o = \mu^*$ and $\Omega_{p,t} = \Omega_{p,t}^*$. With the initial conditions $C_+^o(0) = 0$ and $C_-^o(0) = 1$, we find the solutions

$$C_+^o = \frac{i\Omega_p}{\Omega} \sin\Omega t \exp[-(i\Delta_p + \gamma)t/2] \quad (7a)$$

$$C_-^o = (\cos\Omega t + \frac{\gamma - i\Delta_p}{2\Omega} \sin\Omega t) e^{(i\Delta_p - \gamma)t/2} , \quad (7b)$$

where we have defined the complex Rabi frequency

$$\Omega = \frac{1}{2} [(\Delta_p + i\gamma)^2 + 4\Omega_p^2]^{1/2} \quad (8)$$

The real part of the Rabi frequency is given by

$$\Omega_R = [(\sqrt{x^2 + y^2} + x)/8]^{1/2} , \quad (9a)$$

which determines the oscillation frequency, and the imaginary part by

$$\Omega_i = -[(\sqrt{x^2 + y^2} - x)/8]^{1/2} , \quad (9b)$$

which determines the envelope of the oscillation. In (9) we have defined $x = \Delta_p^2 + 4\Omega_p^2 - \gamma^2$ and $y = 2\Delta_p \gamma$, with $\Delta_p < 0$.

The solutions up to first-order perturbation can be obtained by inserting the zeroth-order solutions (7) into the right-hand side of (5). The results are

$$C_{\pm}(t) = C_{\pm}^0(t) + \delta C_{\pm}(t) \pm A_{\pm} \quad (10)$$

$$\delta C_{-} = \frac{\Omega_t \Omega_p}{\Omega} \frac{\exp[i\Delta_t t - \frac{1}{2}(i\Delta_p + \gamma)t]}{[\frac{1}{2}(i\Delta_p + \gamma) - i\Delta_t]^2 + \Omega^2} \times [(i\Delta_p + \gamma) - i\Delta_t] \sin\Omega t + \Omega \cos\Omega t \quad (11a)$$

$$\delta C_{+} = \frac{i\Omega_t}{2\Omega} \frac{\exp[-i\Delta_t t + \frac{1}{2}(i\Delta_p - \gamma)t]}{[i\Delta_t - \frac{1}{2}(i\Delta_p - \gamma)]^2 + \Omega^2} \times [(\Delta_p + i\gamma)^2 + 2\Omega_p^2 + i\Delta_t(i\Delta_p - \gamma)] \sin\Omega t - 2\Omega[i(\Delta_t - \Delta_p) + \gamma] \cos\Omega t \quad (11b)$$

The integration constants A_{\pm} in (10) are determined by the initial conditions.

The expectation value of the dipole moment of a virtual exciton is

$$\mu(t) = \mu_0 \langle S^-(t) \rangle e^{-i\omega_x t} = \mu_0 C_-^* C_+ e^{i\omega_x t} = \mu_0 (C_-^{0*} + \delta C_-^* + A_-^*)(C_+^0 + \delta C_+ + A_+) e^{-i\omega_x t} \quad (12)$$

It is evident that the dipole moment induced by the test beam is given by

$$\delta\mu(t) = \mu_0 (C_-^{0*} \delta C_+ + C_+^0 \delta C_-^*) e^{-i\omega_x t} \quad (13)$$

Therefore, the nonlinear optical susceptibility experienced by the test beam is simply

$$\begin{aligned} \chi_t &= n\delta\mu(t)/(E_t e^{i\omega_t t}) \\ &= in \frac{|\mu_0|^2}{2\hbar} e^{-\gamma t} \left\{ \frac{1}{2\Omega z_1} [\cos\Omega^* t + i \frac{\Delta_p - i\gamma}{2\Omega^*} \sin\Omega^* t] \right. \\ &\quad \times [(\Delta_p + i\gamma)^2 + 2\Omega_p^2 - \Delta_t(\Delta_p + i\gamma)] \sin\Omega t \\ &\quad - 2\Omega[i(\Delta_t - \Delta_p) + \gamma] \cos\Omega t \\ &\quad \left. - \frac{\Omega_p^2}{|\Omega|^2 z_2} \sin\Omega t [(i\Delta_p - \gamma) - i\Delta_t] \sin\Omega^* t + \Omega^* \cos\Omega^* t \right\} \quad (14) \end{aligned}$$

where

$$z_1 = \left[\frac{1}{2}(i\Delta_p - \gamma) - i\Delta_t \right]^2 + \Omega^2 \quad (15a)$$

$$z_2 = \left[\frac{1}{2}(i\Delta_p - \gamma) - i\Delta_t \right]^2 + \Omega^{*2} \quad (15b)$$

We have introduced in (14) the optically-induced transient virtual exciton density n which may be assumed to obey the rate equation²³

$$\frac{dn}{dt} = \eta \alpha I_p / (\hbar \omega_p) - \gamma n, \quad (16)$$

where η denotes the quantum efficiency of virtual exciton generation per absorbed photon, $I_p = E_p^2 n_0 / 8\pi$ is the pumping intensity with n_0 as the linear refractive index, and α represents the absorption coefficient which is assumed to be a constant in the present case. In practice, however, α may be related to the density n to some extent.

Equation (16) can be solved by a solution of the form

$$n = n_s (1 - e^{-\gamma t}), \quad (17)$$

where $n_s = \eta \alpha I_p / (\hbar \omega_p)$. Once again, the initial condition $n(0) = 0$ is already assumed. The transient behavior of the nonlinear optical susceptibility χ_t is calculated numerically as a function of time and the detuning Δ_t . The results are presented and discussed in the next section.

2.2 Optical nutation and hole burning

In our numerical study, we consider PTS and use the unit $\hbar = 1$. Other parameters are chosen as¹²⁻¹⁴ $\Delta_p = 0.1$ eV, $\Omega_p = 0.02$ eV, $\gamma = 0.01$ eV, $\frac{1}{2}|\mu|^2 n_s = \frac{1}{40\pi}$ eV and $\omega_t = 2$ eV. The real and imaginary parts of χ_t are computed separately. Figure 1 shows snapshots at different instants of time of the variation of χ_t with the detuning Δ_t in the unit of eV. The variation with time for fixed detuning is shown in Fig. 2.

It is clearly seen from Fig. 1 that both the differential absorption and refraction are direct effects of the virtual excitons induced by the pumping field. Nonzero susceptibility starts to show up as $|\Delta_t| \rightarrow 0.1$ and approaches its maximum as $\Delta_t \rightarrow 0$, where the test field is at resonance with the exciton. This indicates that there is energy transfer from the pump field to the test field via virtual excitons, in qualitative agreement with experiments.^{12, 13}

Optical nutation can be observed in polymers in the ultrashort time regime when virtual excitons are generated by a pump field tuned below the exciton resonance. This situation can be seen from Fig. 2. It is also clearly shown that the oscillation centers of both $\text{Re}\chi_t$ and $\text{Im}\chi_t$ change with time. This is quite different from nutation phenomena predicted in Refs. 21 and 23 for semiconductors, where the oscillation amplitude decreases monotonically like damped oscillators.

Since it takes time for the pump field to build up enough virtual excitons and since the virtual excitons are short-lived, their effects are appreciable and stable most of the time during the pulse (on the order of picoseconds). The response time can be anywhere between 10 ~ 100 femtoseconds according to our calculation. Both

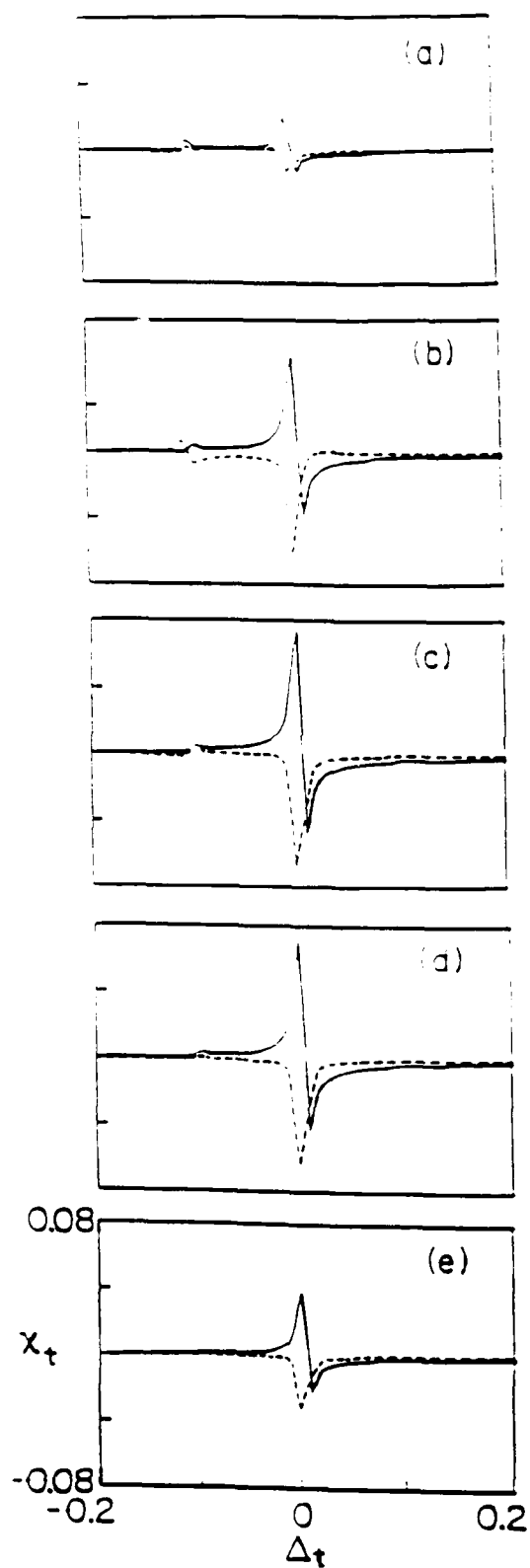


Figure 1. Snapshots of the real (solid lines) and imaginary (dashed line) parts of the nonlinear susceptibility at various times: (a) $t = 25 \text{ eV}^{-1}$, (b) $t = 100 \text{ eV}^{-1}$, (c) $t = 250 \text{ eV}^{-1}$, (d) $t = 500 \text{ eV}^{-1}$ and (e) $t = 1500 \text{ eV}^{-1}$.

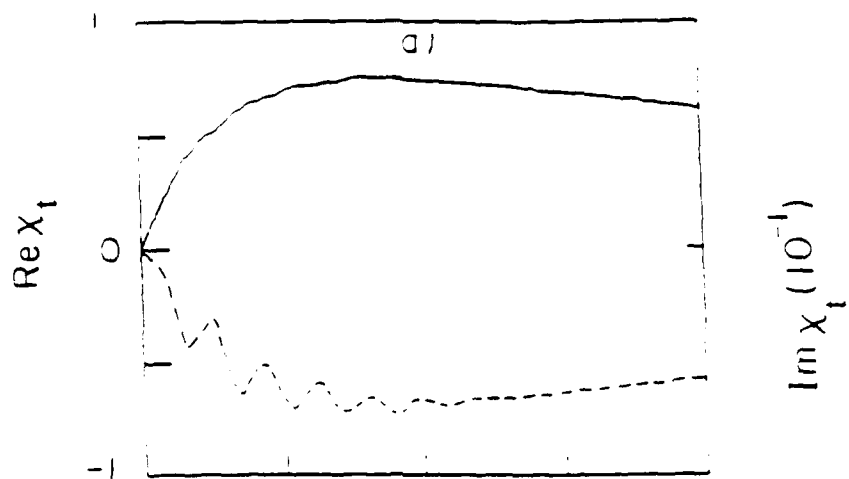
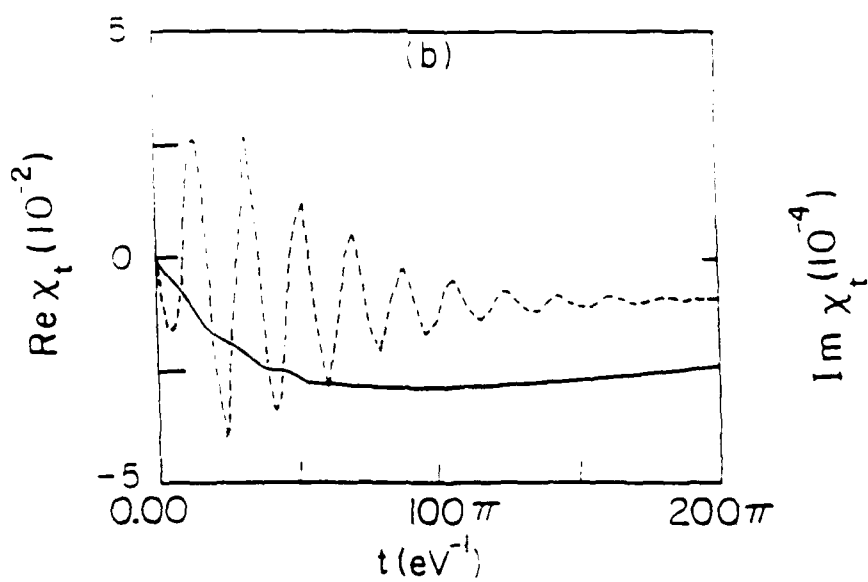


Figure 2. Real (solid lines) and imaginary (dashed lines) parts of χ_t versus time: (a) $\Delta_t = 0$ and (b) $\Delta_t = 0.1$ eV.



the absorption and refraction parts show up almost right away and taper off rather slowly after reaching their peaks.

In addition, we can also observe from these results that increasing the intensity of the pump field leads to a deeper and wider hole in the absorption spectrum, which is qualitatively in agreement with the steady-state results of Ref. 10. What seems to be more interesting is that we find a blue shift of 0.005 eV in our calculation, even though we have not considered the effect of phonons. A more careful treatment with phonon mediation included can be found in Ref. 22. This is the electronic state optical Stark shift observed in a recent experiment¹⁴ in which the differential transmission of polydiacetylene-toluene sulfonate has been measured by means of coherent inverse Raman spectroscopy. It is demonstrated in Ref. 14 that this ac Stark effect is important in the determination of the nonlinear optical response of PTS even in the small signal limit. The effect is reproduced theoretically in Ref. 14 by including an extra term to modify the exciton resonance frequency in the simple model of Schmitt-Rink et al.¹³ However, the treatment in both Refs. 13 and 14 assumes a steady-state solution, while the experimental conditions are more transient-like.

3. PHONON-MEDIATED OPTICAL TRISTABILITY IN PTS

We now turn our attention to a special case in which a sample of PTS is placed inside an optical cavity, and investigate its response to a laser beam directed into the cavity. In particular, we look for the dependence of the output field intensity on the intensity of the incident laser field. To describe our system, we assume the model Hamiltonian of Ref. 13 with an extra term representing the cavity photon-exciton interaction. Thus

$$H = \Omega a^\dagger a + \omega_{ph} b^\dagger b + \omega_{ex} c^\dagger c + \lambda c^\dagger c (b + b^\dagger) + ig(a^\dagger c - c^\dagger a) + i(a^\dagger E e^{-i\Omega t} - a E^* e^{i\Omega t}) \quad (18)$$

where $a^\dagger(a)$, $b^\dagger(b)$ and $c^\dagger(c)$ are the creation (annihilation) operators for the cavity field, phonon and exciton, respectively, with corresponding frequencies Ω , ω_{ph} and ω_{ex} . The phonon-exciton coupling constant is denoted by λ , and the exciton-field coupling constant by g . The coherent driving field is assumed to have the same frequency Ω , and its amplitude is E whose spatial dependence is ignored completely. For simplicity, we have included only one of the phonon modes²⁴ which couples most strongly to the exciton, with momentum dependence neglected.¹³

In the usual zero-fluctuation limit, quantum mechanical operators can be replaced by their expectation values $\alpha = \langle a \rangle$, $\beta = \langle b \rangle$, $\eta = \langle c \rangle$ and $n_c = \langle c^\dagger c \rangle$, and in the limit of high Q cavity, the phonon and exciton variables are eliminated adiabatically.²⁵ Without going into the details which will be published elsewhere,²⁶ we discuss only briefly the results here. The equation of motion for the field variable, expressed in the unit such that $\gamma_{ex} = 1$, takes the form

$$\frac{\partial}{\partial \tau} \alpha_c = \sqrt{2g^2} E - \alpha_c f(|\alpha_c|^2) \quad (19)$$

where the function f is defined as

$$f(x) = 1 + \left(\frac{g^2}{\kappa}\right) \frac{1 - i[\Delta - \lambda_p n_c(x)]}{1 + [\Delta - \lambda_p n_c(x)]^2} \quad (20)$$

In Eq. (19), we have introduced for convenience the phenomenological damping rates κ , γ_{ph} and γ_{ex} and dimensionless variables $\tau = \kappa t$, $E = E/\kappa$ and $\alpha_c = \bar{E}g\alpha_0$. The field intensity is expressed as $I = |\alpha_c|^2$ and the detuning as $\Delta = \omega_{ex} - \Omega$.

In the steady state, we set $\alpha_c = \alpha_s = \text{constant}$ in (19) and (20). The steady-state cavity field intensity can then be found straightforwardly as

$$I_s = \frac{1 + Y_s^2(I_{in})}{(1 + g^2/\kappa)^2 + Y_s^2(I_{in})} I_{in} \quad (21)$$

where we have defined the driving field intensity $I_{in} = 2g^2|E|^2$ and $Y_s(I_{in}) = \Delta - \lambda_p n_s(I_{in})$ with $\lambda_p = 2\lambda^2/\omega_{ph}$. It can be shown from (21) that the steady-state virtual exciton n_s satisfies the equation

$$n_s^3 - 2(\Delta/\lambda_p)n_s^2 + [\Delta^2 + (1 + g^2/\kappa)^2]\lambda_p^{-2}n_s - I_{in}\lambda_p^{-2} = 0 \quad (22)$$

When the conditions are such that Eq. (22) has three distinct roots, there will be three output I_s for each given I_{in} . As has been discussed in Ref. 26, the conditions of multi-real roots of (22) require that tristability occurs only when the cavity field is tuned sufficiently below the exciton resonance and when there exists phonon-exciton coupling. This clearly shows that the tristability is mediated by phonons.

Numerical calculation of Eq. (21) has been performed for PTS for which^{13,14} $\omega_{ph} = 5.16$ and $\lambda = 2.0$. The results are plotted in Fig. 3 for various choices of Δ and g^2/κ . It is found that all the multiple solutions are stable except for those around the turning points A and B in Figs. 3(b) and 3(c). As I_{in} increases continuously, passes the turning point and then decreases, the output I_s will follow a reversed hysteresis loop. The middle branch can only be reached if I_{in} reverses its course of change at the turning point.

It is emphasized that the dispersion effect plays a crucial role in this kind of tristability. When I_{in} increases beyond a certain level, n_s becomes large enough that the lattice relaxation is sufficiently high for the refractive index to deviate from the cavity resonance. Consequently, the cavity field switches to the lower branch. On the other hand, such tristability cannot occur when the Q-value of the optical cavity is excessively high. As a final remark, we remark that this tristability is found by considering only one phonon mode in PTS. It is therefore expected to find optical multistability if more phonon modes are included.

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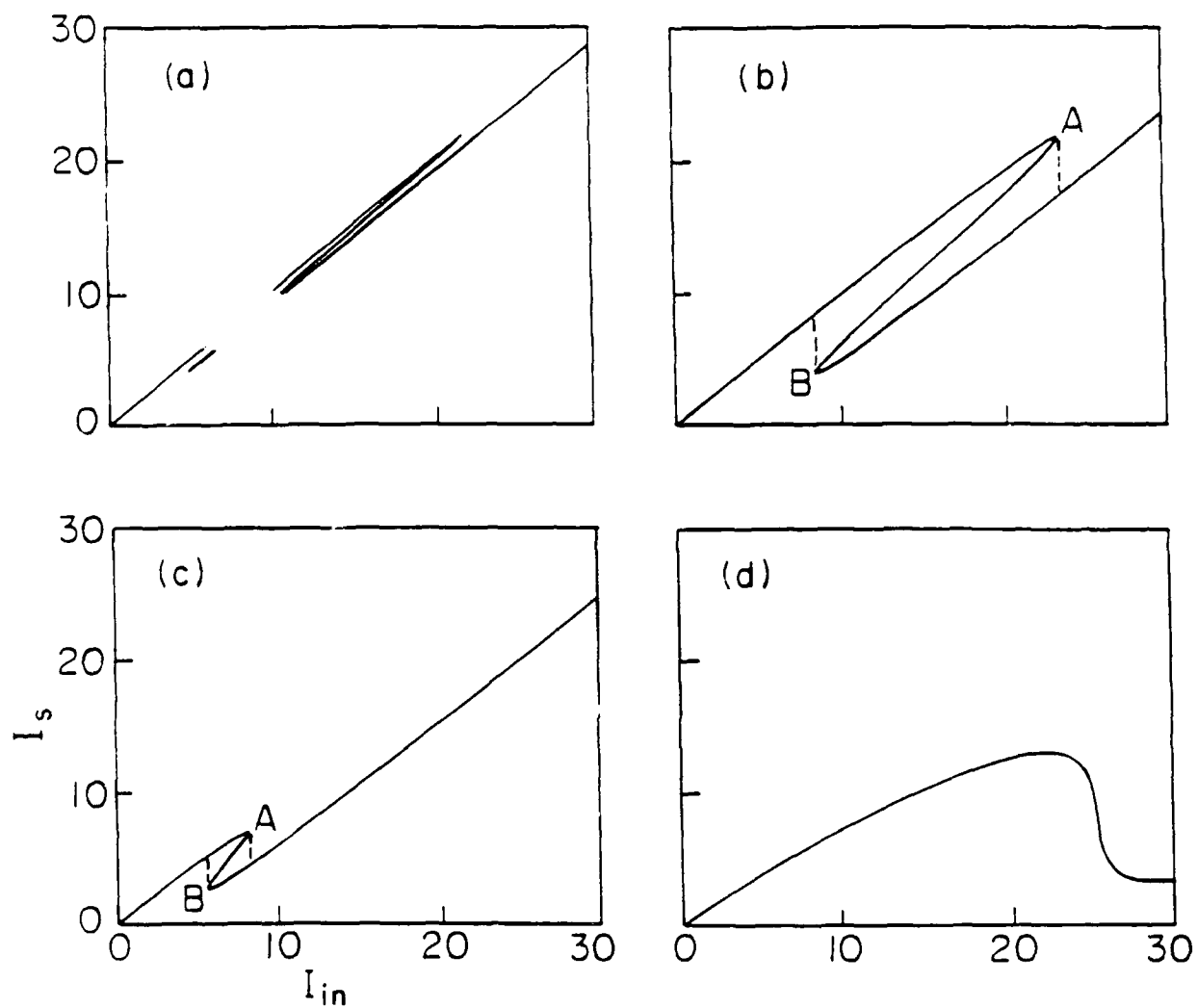


Figure 3. Intensity of the cavity field I_s versus the input field intensity I_{in} : (a) $\Delta = 6, g^2/\kappa = 0.1$; (b) $\Delta = 6, g^2/\kappa = 0.5$; (c) $\Delta = 4, g^2/\kappa = 0.5$; (d) $\Delta = 5, g^2/\kappa = 2$. The dashed lines in (b) and (c) are hypothetical and drawn only to complete the reversed hysteresis loop.

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